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REMARKS

Applicants would like to thank Examiner Wills for the courtesy extended to the undersigned, Dorothy P. Whelan, and Dr. Kevin Eberman during the telephone interview conducted on May 20, 2004. During the interview, the outstanding rejection of claims 1-19 under 35 U.S.C. §112, first paragraph, for failing to satisfy the written description requirement was discussed. Independent claim 1 had been amended in response to the First Office Action to define x as being (a) greater than 0 but less than 0.5 (i.e., $0 < x < 0.5$) or (b) greater than 0.5 but less than 1 (i.e., $0.5 < x < 1$). Claims 1, 3, 8, and 13 had been amended to recite that when M^1, M^2, M^3, M^4, or M^5 includes Ni, Co, or a combination thereof, all of the Ni, Co, and Mn included in the composition have oxidation states in air of +2, +3, and +4, respectively.

During the interview, the undersigned and Dr. Eberman explained that the amendment to claim 1 defining the range for x found support in claims 3 and 13, both of which depended on claim 1. As dependent claims, these two claims necessarily are narrower than independent claim 1. Claim 3 requires that $x = (2-y)/3$ <u>and</u> that $0 < y < 0.5$. Treating this requirement as an algebraic equation and solving for x, it is evident that $x = 0.5$ only when $y = 0.5$. However, claim 3 excludes this possibility by defining y as being less than (but never equal to) 0.5. Accordingly, claim 3 provides support for the language in claim 1 calling for $0.5 < x < 1$. The Examiner agreed.

Claim 13 requires x to be equal to y, and then goes on to define y as $0 < y < 0.5$. Substituting x for y, it is evident that claim 13 supports the language in claim 1 calling for $0 < x < 0.5$. The Examiner also agreed.

Turning to the amendments to claims 1, 3, 8, and 13 relating to oxidation states, the undersigned and Dr. Eberman explained that the compositions covered in Applicants' claims are ceramic oxides. Dr. Eberman further explained that electrical neutrality is an inherent condition of every ceramic oxide material, and offered to provide literature support for this proposition. Accordingly, an excerpt from a seminal text in the ceramics area, *Introduction to Ceramics*, by Kingery, Bowen, and Uhlmann (1976), is included with this reply. The reference states (at page 60:

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A stable structure must be electrically neutral not only on a macroscopic scale but also at the atomic level.

To satisfy the condition of electrical neutrality, the sum of the products of the stoichiometric subscripts and oxidation states of the elements forming the ceramic oxide must equal zero.

Applying the requirement of charge neutrality to the compositions that are the subject of Applicants' claims, we refer the Examiner to the following passage in Applicants' specification:
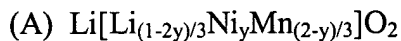
> [T]he particular metal elements are selected such that they exhibit appropriate oxidation states in air at the desired synthesis temperature. Conversely, the synthesis temperature may be adjusted so that a particular metal element exists in a desired oxidation state in air at that temperature.
>
> In general, examples of suitable metal elements for inclusion in the cathode composition include Ni, Co, Fe, Cu, Li, Zn, V, and combinations thereof. Particularly preferred cathode compositions are those having the following formulae:
>
> $Li[Li_{(1-2y)/3}Ni_yMn_{(2-y)/3}]O_2$;
> $Li[Li_{(1-y)/3}Co_yMn_{(2-2y)/3}]O_2$; and
> $Li[Ni_yCo_{1-2y}Mn_y]O_2$.

In the case of Ni, Co, and Mn, this passage requires oxidation states of +2, +3, and +4, respectively, in order to achieve electrical neutrality, as shown by working out the generalized charge balance equation for each of the three formulae. The charge balance equation is written as the sum of the oxidation states of the individual elements multiplied by their stoichiometric subscripts. The total must equal zero to satisfy the requirement of electrical neutrality.

The oxidation state of Li must be +1 and the oxidation state of O must be −2. If we start by saying that a, b, and c are the unknown oxidation states for the Ni, Co, and Mn, respectively, in the three formulae quoted above, we can analyze each equation as follows (where the number in brackets represents the oxidation state of the particular element):

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(A) $Li[Li_{(1-2y)/3}Ni_yMn_{(2-y)/3}]O_2$

$1[1.0] + ((1-2y)/3) [1.0] + y[a] + ((2-y)/3)[c] + 2[-2] = 0$

When multiplied out, this equation reduces to:
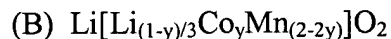
$1 + (1/3) - y(2/3) + ya + (2/3)c - y(1/3)c - 4 = 0$

Collecting the terms yields:

$y(a - (1/3)c - (2/3)) = (8/3) - (2/3)c$

Further simplifying the equation yields:

$y(3a - c - 2) = 8 - 2c$

It is only when $(3a - c - 2) = 0$ and $(8 - 2c) = 0$ that this equation is satisfied for all values of y. This means that c = 4 and a = 2. Thus, electrical neutrality is obtained only when Mn is in its +4 state (i.e., c = 4) and Ni is in its +2 state (i.e., a = 2).

(B) $Li[Li_{(1-y)/3}Co_yMn_{(2-2y)}]O_2$

$1[1.0] + ((1-y)/3) [1.0] + y[b] + ((2-2y)/3)[c] + 2[-2] = 0$

When multiplied out, this equation reduces to:
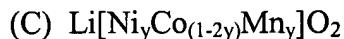
$1 + (1/3) - y(1/3) + yb + (2/3)c - y(2/3)c - 4 = 0$

Collecting the terms yields:

$y(b - (2/3)c - (1/3)) = (8/3) - (2/3)c$

Further simplifying the equation yields:

$y(3b - 2c - 1) = 8 - 2c$

It is only when $(3b - 2c - 1) = 0$ and $(8 - 2c) = 0$ that this equation is satisfied for all values of y. This means that $c = 4$ and $b = 3$. Thus, charge neutrality is obtained only when Mn is in its +4 state (i.e., $c = 4$) and Co is in its +3 state (i.e., $b = 3$).

(C) $Li[Ni_yCo_{(1-2y)}Mn_y]O_2$

$1[1.0] + y[a] + (1-2y)[b] + y[c] + 2[-2] = 0$

When multiplied out, this equation reduces to:

$1 + ya + b - y2b + yc - 4 = 0$

Collecting the terms yields:

$y(a - 2b + c) = 3 - b$

It is only when $(a - 2b + c) = 0$ and $(3 - b) = 0$ that this equation is satisfied for all values of y. This means that $b = 3$. For $(a + c)$, we have: $a - 2b + c = 0$, which yields $a + c = 6$. Taking $a = 2$ from Formula (A) yields $c = 4$. Similarly, taking $c = 4$ from Formula (B) yields $a = 2$. Thus, charge neutrality is obtained only when Mn is in its +4 state (i.e., $c = 4$), Co is in its +3 state (i.e., $b = 3$), and Ni is in its +2 state.

The specification, therefore, provides support for the oxidation states recited in the claims.

The Examiner questioned whether the four prior art references over which the claims had been rejected also described transition metal oxides in which the transition metal elements had the oxidation states recited in the amended claims. None of these references describes oxides having the oxidation states recited in Applicants' amended claims in combination with the range of values for x also recited in Applicants' amended claims. Applicants have discovered that this combination yields cathode compositions having improved stability, as reflected in their ability to resist transforming into a spinel crystal structure when incorporated in a lithium-ion battery

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and cycled under the conditions set forth in claim 1. We will address each cited reference individually.

(1) Nitta. The only Mn-containing embodiments described in Nitta lack Ni in its +2 oxidation state and Mn in its +4 oxidation state, as the claims require. Specifically, Nitta describes a series of compounds having the formula $Li_yNi_{1-x}Mn_xO_2$ where y = 0.1, 1.0, 1.1, 1.3, or 1.5, and x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, or 0.7 (see Figs. 13-14 and Examples 1-2). Of these compounds, there are only six whose stoichiometry would satisfy the formula set forth in Applicants' amended claims 1-2 and 19 (i.e., $Li[M^1_{(1-x)}Mn_x]O_2$, where (a) 0 < x < 0.5 or (b) 0.5 < x < 1:

(a) $LiNi_{0.9}Mn_{0.1}O_2$;

(b) $LiNi_{0.8}Mn_{0.2}O_2$;

(c) $LiNi_{0.7}Mn_{0.3}O_2$;

(d) $LiNi_{0.6}Mn_{0.4}O_2$;

(e) $LiNi_{0.4}Mn_{0.6}O_2$;

(f) $LiNi_{0.3}Mn_{0.7}O_2$.

However, none of these compounds includes all of the Ni in its +2 oxidation state and all of the Mn in its +4 oxidation state, as further required by Applicants' claims, because none of these compounds would achieve electrical neutrality with these oxidation states, as the following calculations demonstrate:

(a)   Lithium = 1.0 [+1] = 1.0;

            Nickel = 0.9 [+2] = 1.8;

            Manganese = 0.1 [+4] = 0.4;

            Oxygen = 2.0 [-2] = -4.0;

            1.0 + 1.8 + 0.4 − 4.0 = -0.8.

(b)   Lithium = 1.0 [+1] = 1.0;

Nickel = 0.8 [+2] = 1.6;

Manganese = 0.2 [+4] = 0.8;

Oxygen = 2.0 [-2] = -4.0;

1.0 + 1.6 + 0.8 − 4.0 = -0.6.

(c)   Lithium = 1.0 [+1] = 1.0;

Nickel = 0.7 [+2] = 1.4;

Manganese = 0.3 [+4] = 1.2;

Oxygen = 2.0 [-2] = -4.0;

1.0 + 1.4 + 1.2 − 4.0 = -0.4.

(d)   Lithium = 1.0 [+1] = 1.0;

Nickel = 0.6 [+2] = 1.2;

Manganese = 0.4 [+4] = 1.6;

Oxygen = 2.0 [-2] = -4.0;

1.0 + 1.2 + 1.6 − 4.0 = -0.2.

(e)   Lithium = 1.0 [+1] = 1.0;

Nickel = 0.4 [+2] = 0.8;

Manganese = 0.6 [+4] = 2.4;

Oxygen = 2.0 [-2] = -4.0;

1.0 + 0.8 + 2.4 − 4.0 = 0.2.

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(f)   Lithium = 1.0 [+1] = 1.0;

Nickel = 0.3 [+2] = 0.6;

Manganese = 0.7 [+4] = 2.8;

Oxygen = 2.0 [-2] = -4.0;

1.0 + 0.6 + 2.8 − 4.0 = 0.4.

The ceramic oxides described in Nitta, therefore, cannot achieve electrical neutrality when Ni and Mn have oxidation states of +2 and +4, respectively, as Applicants' claims require. On the contrary, in order to achieve the electrical neutrality required of all ceramic oxides, the Ni and Mn included in Nitta's compositions must have different oxidation states. Nitta's compositions are thus distinct from the compositions that are the subject of Applicants' claims.

(2) Paulsen. Paulsen proposes cathode compositions that fall within one of two broad generic formulae: $Li[Li_xCo_y(Mn_zNi_{1-z})_{1-x-y}]O_2$ or $Li[Li_xCo_y(Mn_2Ni_{1-z})_{1-x-y}]O_2$, where x = 0 - 0.16; y = 0.1 - 0.3; and z = 0.4 − 0.65. While it might theoretically be possible to select appropriate values of a, b, and c such that Ni, Co, and Mn have oxidation states of +2, +3, and +4, respectively, in air, Paulsen provides no guidance whatsoever as to how to select the particular combinations necessary in order to achieve this result from among the multitude of possible combinations. Moreover, the only working examples that Paulsen provides lack Ni, Co, and Mn in these oxidation states (where the charge value was calculated as described above in the case of Nitta):

Example 2A: $Li[Li_{0.095}Co_{0.046}Ni_{0.434}Mn_{0.424}]O_2$

Charge = -0.200

Example 2B: $Li[Li_{0.134}Co_{0.044}Ni_{0.416}Mn_{0.406}]O_2$

Charge = -0.278

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Example 2C: $Li[Li_{0.083}Co_{0.093}Ni_{0.413}Mn_{0.412}]O_2$

Charge = -0.167

Example 2D: $Li[Li_{0.123}Co_{0.089}Ni_{0.395}Mn_{0.394}]O_2$

Charge = -0.247

Example 2E: $Li[Li_{0.064}Co_{0.158}Ni_{0.388}Mn_{0.389}]O_2$

Charge = -0.127

Example 2F: $Li[Li_{0.106}Co_{0.151}Ni_{0.371}Mn_{0.372}]O_2$

Charge = -0.211

Example 4: $Li[Li_{0.048}Co_{0.152}Ni_{0.39}Mn_{0.41}]O_2$

Charge = -0.076

Example 5A: $Li[Li_{0.11}Co_{0.148}Ni_{0.371}Mn_{0.371}]O_2$

Charge = -0.220

Example 5B: $Li[Li_{0.11}Co_{0.142}Ni_{0.365}Mn_{0.383}]O_2$

Charge = -0.202

Example 5C: $Li[Li_{0.09}Co_{0.146}Ni_{0.373}Mn_{0.391}]O_2$

Charge = -0.162

Example 6: $Li[Li_{0.05}Co_{0.14}Ni_{0.37}Mn_{0.37}Al_{0.07}]O_2$

Charge = -0.1 (assuming a +3 oxidation state for Al).

As the calculations demonstrate, electrical neutrality cannot be obtained if all of the Ni, Co, and Mn are in their +2, +3, and +4 oxidation states, respectively, as Applicants' claims require. Therefore, Ni, Co, and Mn must have different oxidation states in Paulsen's compositions. Paulsen's compositions are thus distinct from Applicants' claimed compositions.

(3) Sunagawa. Sunagawa describes compositions having the formula $Li_aCo_bM_cNi_{1-b-c}O_2$, where M may be Mn. As in the case of Paulsen, while it might theoretically be possible to select appropriate values of a, b, and c such that Ni, Co, and Mn have oxidation states of +2, +3, and +4, respectively, in air, Sunagawa provides no guidance whatsoever as to how to select the particular combinations necessary in order to achieve this result from among the multitude of possible combinations. Moreover, the only Mn-containing examples that Sunagawa provides lack Ni, Co, and Mn in these oxidation states (where the charge value was calculated as described above in the case of Nitta):

Example A1: $Li[Co_{0.01}Mn_{0.01}Ni_{0.98}]O_2$
Charge = -0.97.

Example A2: $Li[Co_{0.01}Mn_{0.20}Ni_{0.79}]O_2$
Charge = -0.59.

Example A3: $Li[Co_{0.01}Mn_{0.40}Ni_{0.59}]O_2$
Charge = -0.19.

Example A4: $Li[Co_{0.1}Mn_{0.40}Ni_{0.50}]O_2$
Charge = -0.10.

Example A5: $Li[Co_{0.20}Mn_{0.01}Ni_{0.79}]O_2$
Charge = -0.78.

Example A6: $Li[Co_{0.20}Mn_{0.20}Ni_{0.60}]O_2$

Charge = -0.4.

Example A7: $Li[Co_{0.20}Mn_{0.30}Ni_{0.50}]O_2$

Charge = -0.2.

Example A8: $Li[Co_{0.40}Mn_{0.01}Ni_{0.59}]O_2$

Charge = -0.58.

Example A9: $Li[Co_{0.40}Mn_{0.10}Ni_{0.50}]O_2$

Charge = -0.40.

Example B1: $Li[Co_{0.45}Mn_{0.05}Ni_{0.50}]O_2$

Charge = -0.45.

Example B2: $Li[Co_{0.05}Mn_{0.45}Ni_{0.50}]O_2$

Charge = -0.05.

Example B3: $Li[Co_{0.15}Mn_{0.40}Ni_{0.45}]O_2$

Charge = -0.05.

Example B4: $Li[Co_{0.40}Mn_{0.15}Ni_{0.45}]O_2$

Charge = -0.30.

Example B5: $Li[Co_{0.00}Mn_{0.10}Ni_{0.90}]O_2$

Charge = -0.80.

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Example 4: $Li[Co_{0.30}Mn_{0.10}Ni_{0.60}]O_2$

Charge = -0.50.

As the calculations demonstrate, electrical neutrality cannot be obtained if all of the Ni, Co, and Mn are in their +2, +3, and +4 oxidation states, respectively, as Applicants' claims require. Therefore, Ni, Co, and Mn must have different oxidation states in Sunagawa's compositions. Sunagawa's compositions are thus distinct from Applicants' claimed compositions.

   (4) Yanai. Yanai describes cathode compositions that include a combination of a lithium-containing metal oxide and an aluminum sulfate. None of these compositions falls within the claims of the present application because the presence of the aluminum sulfate means that these compositions are not single phase compositions, as the claims require.

   Yanai does describe, as comparative examples, three compositions that lack the aluminum sulfate but include a lithium metal oxide having Mn in combination with Ni and/or Co. These compositions are set forth in Table 2 of Yanai and have the following compositions:

   (a) $LiNi_{0.5}Mn_{0.5}O_2$;

   (b) $LiCo_{0.5}Mn_{0.5}O_2$;

   (c) $LiNi_{0.4}Co_{0.1}Mn_{0.5}O_2$.

None of these compositions, however, meets the requirements of claims 1-7 and 19 because each contains 0.5 moles of Mn. The claims, in contrast, require the amount of Mn to be either <u>greater than</u> 0.5 (i.e., $0.5 < x < 1$) or <u>less than</u> 0.5 (i.e., $0 < x < 0.5$), but never <u>equal to</u> 0.5.

In conclusion, the amended claims satisfy the written description requirement of 35 U.S.C. §112, first paragraph. Moreover, they distinguish the four cited references, each of which fails to describe transition metal oxides that satisfy the oxidation state and stoichiometry requirements that Applicants have discovered yield superior cathode compositions. Accordingly, Applicants respectfully request that the outstanding rejections be withdrawn.

Please apply any other charges or credits to deposit account 06-1050.

Respectfully submitted,

Date: 5/24/04

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Introduction
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60   **INTRODUCTION TO CERAMICS**

nearest-neighbor interactions. A stable structure must be electrically neutral not only on a macroscopic scale but also at the atomic level. Pauling's *second rule* describes a basis for evaluating local electrical neutrality. We define the *strength* of an ionic bond donated from a cation to an anion as the formal charge on the cation divided by its coordination number. For example, silicon, with valence 4 and tetrahedral coordination, has bond strength $4/4 = 1$; Al^{3+} with octahedral coordination has bond strength $3/6 = 1/2$. (The same considerations are applied regardless of whether all coordinating anions are the same chemical species; the bond strength of Al^{3+} is $1/2$ in both the structure of Al_2O_3, Fig. 14.28, where the six anion neighbors are O^{2-}, and in that of kaolinite, Fig. 2.35, where the anions surrounding Al^{3+} are $4OH^-$ and $2O^{2-}$.) The second rule states that in a stable structure the total strength of the bonds reaching an anion from all surrounding cations should be equal to the charge of the anion. For example, in the Si_2O_7 unit, Fig. 2.22a, two bonds of strength 1 reach the shared oxygen ion from the surrounding silicon ions; the sum of the bonds is thus 2, the valence of the oxygen ion. (Note that this implies that, in a silicate based on Si_2O_7 units, no additional cation may be bonded to this shared oxygen.) Similarly in the structure of spinel $MgAl_2O_4$, Fig. 2.25, each O^{2-} is surrounded by one Mg^{2+} which donates a bond of strength $2/4$ and three Al^{3+} which donate three bonds of strength $3/6$.

Pauling's *third rule* further concerns the linkage of the cation coordination polyhedra. In a stable structure the corners, rather than the edges and especially the faces, of the coordination polyhedra tend to be shared. If an edge is shared, it tends to be shortened. The basis of this rule is again
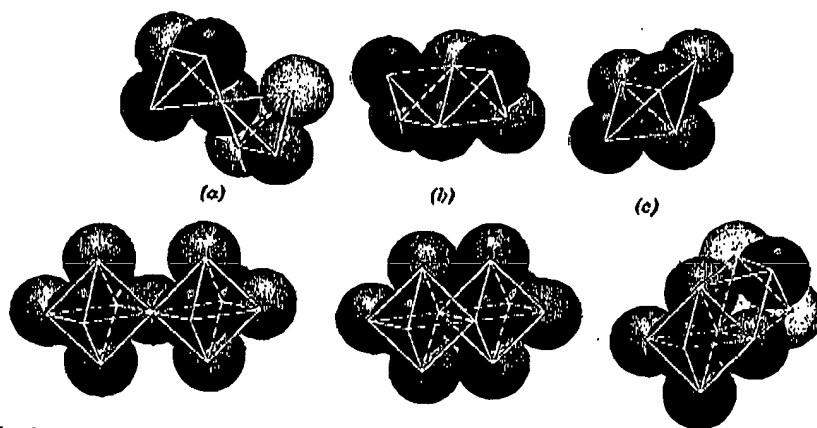

Fig. 2.22. Tetrahedra and octahedra linked by sharing (*a*) corner, (*b*) edge, and (*c*) face (reference 3).